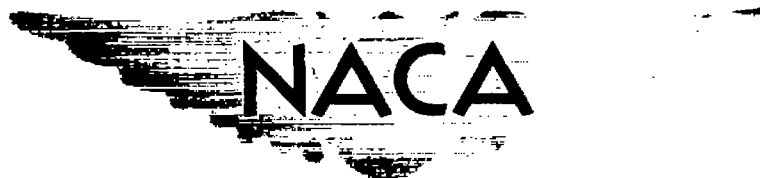


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RESEARCH MEMORANDUM

FLAME VELOCITIES OF PROPANE- AND
ETHYLENE-OXYGEN-NITROGEN
MIXTURES

By Gordon L. Dugger and Dorothy D. Graab

Lewis Flight Propulsion Laboratory
Cleveland, Ohio

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RESEARCH MEMORANDUM

NATIONAL ADVISORY COMMITTEE
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FLAME VELOCITIES OF PROPANE- AND ETHYLENE-

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SUMMARY

The laminar flame velocities of propane and ethylene with various oxygen-nitrogen mixtures were determined from schlieren photographs of Bunsen-type flames. Flame velocity was determined as a function of equivalence ratio for each oxygen-nitrogen mixture at two initial temperatures, 311° and 422° K. The mole fraction of oxygen in the oxygen-nitrogen mixture was varied from 0.166 to 0.496 for propane and from 0.166 to 0.349 for ethylene. The maximum flame velocity with respect to equivalence ratio was found to vary linearly with the mole fraction of oxygen α for each initial temperature T_0 (°K). With the assumption of a common α -intercept at zero flame velocity for both initial temperatures, empirical equations including the effect of α and of T_0 were obtained.

The experimental results for the effect of α are predicted on a relative basis, within approximately 5 to 15 percent, by approximate theoretical equations based on either a thermal mechanism, as presented by Semenov, or an active-particle diffusion mechanism, as presented by Tanford and Pease. Both theories predict a decreasing rate of change in maximum flame velocity with increasing oxygen concentration rather than the linear relation between U_{max} and α observed experimentally. Neither the sum of the effective active-particle concentrations nor the square root of this sum is related linearly to maximum flame velocity for the entire range of α investigated.

INTRODUCTION

A study of the effect of oxygen concentration on the flame velocity of isooctane-oxygen-nitrogen mixtures showed that flame velocity increased linearly with the mole fraction of oxygen in the oxidant mixture $O_2/(O_2 + N_2)$ over the range from 0.210 to 0.496 (reference 1). This linear relation was shown to be at variance with the predictions of the approximate solutions of two theoretical equations, one based on a thermal and the other based on a diffusion mechanism of propagation, either

of which predicted a decreasing rate of change of flame velocity with increasing oxygen concentration. It was desirable to know whether the observed linear augmentation of flame velocity would also occur with other fuels, and whether the trends predicted by the theoretical equations would again differ from those established experimentally. Further, data on the effects of the variables equivalence ratio and initial temperature have been obtained for propane- and ethylene-air mixtures (reference 2), and it was thought desirable to obtain data on the effect of oxygen concentration for these same fuels.

For these reasons, flame-velocity data were obtained for propane- and ethylene-oxygen-nitrogen mixtures for which $O_2/(O_2 + N_2)$ varied from 0.166 to 0.496 and from 0.166 to 0.349, respectively. Flame velocity was determined as a function of equivalence ratio at each $O_2/(O_2 + N_2)$ at both 311° and 422° K. Measurements were taken from schlieren photographs of the Bunsen cone, and calculations were made by the total-area method. The maximum flame velocities (with respect to equivalence ratio) are compared with the values predicted by the Semenov bimolecular equation (thermal mechanism, reference 3) and the Tanford-Pease equation (diffusion mechanism, reference 4) on a relative basis.

SYMBOLS

The following symbols are used in this report:

A, B, C	empirical constants
A_F	axial cross-sectional area of flame cone, cm^2
a	fuel concentration, molecules/ cm^3
B_i	term near unity arising from recombination of i^{th} radical
b	oxygen concentration, molecules/ cm^3
C_p	molar heat capacity at constant pressure, cal/(mole)(°K)
c_p	specific heat, cal/(g)(°K)
$\overline{c_p}$	mean specific heat, T_0 to T_f , cal/(g)(°K)
D	diffusion coefficient, cm^2/sec
D_m	diffusion coefficient at mean combustion zone temperature, cm^2/sec

E	activation energy, kcal/g-mole
exp	base of Napierian logarithmic system raised to power in parenthesis following exp
h	height of cone, cm
k_a	weighted mean k_i for active particles (H, OH, and O), each reacting with fuel molecules, $\text{cm}^3/(\text{molecule})(\text{sec})$
k_i	specific rate constant for reaction between i^{th} radical and combustible material, $\text{cm}^3/(\text{molecule})(\text{sec})$
L_m	total concentration of gas at mean combustion zone temperature, molecules/ cm^3
l	height of generating curve, cm
M	molecular weight
m	empirical constant
N	mole fraction
n	total number of molecules of H_2O and CO_2 in products per molecule of fuel by stoichiometric relation
n_r/n_p	moles of reactants per moles of products from stoichiometric equation
P	steric factor used herein as arbitrary constant, from $k = PZ \exp(-E/RT)$
p	total pressure, dynes/ cm^2
p_i	partial pressure or mole fraction of i^{th} active particle in burned gas
Q	mole fraction of potential combustion product in unburned gas
Q'	mole fraction of combustible in unburned gas
R	gas constant, kcal/(g-mole)($^{\circ}\text{K}$) or ergs/(g-mole)($^{\circ}\text{K}$)
S	lateral surface area, cm^2
T	absolute temperature, $^{\circ}\text{K}$

U	flame velocity, cm/sec
U_{\max}	maximum flame velocity, varying ϕ at constant α and T_0 , cm/sec
Z	collision number, molecular collisions/(sec)(one molecule of each type/cm ³)
α	mole fraction of oxygen in oxygen-nitrogen mixture
α_x	empirical constant
η	viscosity of mixture, poise
θ_m	ratio of mean combustion zone temperature to initial temperature
λ	thermal conductivity, cal/(cm ²)(sec)(°K/cm)
ρ	density of mixture, g/cm ³
σ	collision diameter, cm
ϕ	equivalence ratio, fraction of stoichiometric fuel-oxygen ratio

Subscripts:

0	initial condition
1,2	reactants in bimolecular reaction
eff	effective mean reactant concentration
f	condition at flame temperature
i	i th active particle
j	j th component in mixture

EXPERIMENTAL PROCEDURE

Apparatus. - The apparatus used in this investigation is diagrammatically illustrated in figure 1. The experimental procedure was essentially the same as that reported in reference 1. The fuel and the oxygen-nitrogen mixture were metered through critical flow orifices I (fig. 1) and mixed at point K. The mixture passed through a small heated plenum chamber M and into a copper tube that passed through the oil jacket of the burner P and was connected to the base of the burner

2750 tube. The mixture acquired the temperature of the circulating oil and was burned as a Bunsen-type flame at atmospheric pressure. The circulating oil, and hence the gas mixture, was maintained at either 311° or 422° K $\pm 1^{\circ}$ K by electric heaters.

The burner tube shown in figure 1 was a 120-centimeter length of brass tubing having an inside diameter of 1.256 centimeters. In order to avoid flashback of the higher percentage oxygen mixtures, an insert tube with a lip at the top was soft-soldered to the stationary tube. The insert tubes were 60 centimeters long and had inside diameters of 0.297, 0.465, 0.617, and 0.838 centimeter.

Optical system. - A two-mirror (z-type) schlieren system (reference 5) with a horizontal knife edge was used. A 25-watt concentrated arc lamp served as the light source. The camera lens gave a 1:1 image size, and the exposure time was 1/50 second.

Flame-velocity measurement. - Flame velocities were determined from the schlieren photographs by the total-area method, wherein the average normal flame velocity is equal to the volume rate of flow of the unburned mixture divided by the surface area of the cone formed by the combustion zone. This surface area S was determined by the approximate relation for the cone-like surfaces of revolution:

$$S = \pi A_F l / h \quad (1)$$

Measurements were based on the outside edge of the schlieren image. In order to aid in determining the maximum flame velocities, data scatter was reduced by the method described in reference 2 (an averaging method which diminishes errors in measurements for equation (1)); however, essentially the same results would have been obtained without this technique.

Fuels and oxygen-nitrogen mixtures. - The minimum purities claimed by the supplier of the propane and ethylene were 96 and 99.5 mole percent, respectively. Oxygen and nitrogen were obtained premixed and were analyzed by the supplier to ± 0.1 mole percent. For the 21-percent mixture, compressed air containing approximately 0.4 mole percent water was used.

EXPERIMENTAL RESULTS

In figure 2(a), flame velocity is plotted against the equivalence ratio (fraction of stoichiometric fuel-oxygen ratio) for propane-oxygen-nitrogen mixtures for α values from 0.166 to 0.496 at an initial temperature of 311° K and atmospheric pressure. Similar data are presented

for propane-oxygen-nitrogen mixtures at 422° K in figure 2(b) and for ethylene-oxygen-nitrogen mixtures over a range of α from 0.166 to 0.349 at each of the two temperatures in figures 2(c) and 2(d). These curves exhibit flame velocity maximums at equivalence ratios between 1.00 and 1.06 for propane mixtures and between 1.08 and 1.16 for ethylene mixtures. These maximum flame velocities are presented in table I along with those from some additional curves.

In figure 3, maximum flame velocities from figure 2 are plotted against α for initial temperatures of 311° and 422° K. Linear relations are observed for each fuel at each temperature; that is,

$$U_{\max} = A\alpha - B \quad (2)$$

The constants A and B, determined for each temperature by the method of least squares, are presented in table II. The constants for isooctane (2,2,4-trimethylpentane) from reference 1 are included for comparison.

An empirical equation for each fuel including the effects of both initial temperature T_0 and α was derived by the method described in reference 1, on the assumption that the variation of the α -intercept with temperature would be negligible for a small temperature range:

$$U_{\max} = C T_0^m (\alpha - \alpha_x) \quad (3)$$

The constants C and m were also determined by the method of least squares by forcing the linear equations of type (2) to go through α_x , the average intercept for the two temperatures; these constants are presented in table II. The maximum flame velocities computed from equations (2) and (3) are included in table I.

THEORETICAL PREDICTIONS

Thermal mechanism. - An approximate theoretical equation for flame velocity based primarily on a mechanism of heat conduction from the flame zone into the unburned gases is presented by Semenov in reference 3. For a bimolecular reaction between fuel molecules and oxygen molecules, the equation may be written in the form (see reference 3, pp. 31, 45, 48, and 49):

$$U = \sqrt{\frac{2\lambda PZ a_{\text{eff}} b_{\text{eff}}}{a_0 \rho_0 \bar{c}_p (T_f - T_0)} \left(\frac{\lambda}{c_p^D \rho}\right)_f^2 \left(\frac{n_r}{n_p}\right)^2 \frac{RT_f^2}{E} \exp\left(-\frac{E}{RT_f}\right)} \quad (4)$$

where, for stoichiometric or fuel-rich mixtures ($\phi \geq 1$), a_{eff} and b_{eff} are computed by:

$$\left. \begin{aligned} a_{\text{eff}} &= a_0 \frac{T_0}{T_f} \left[1 - \frac{1}{\phi} \left(1 - \frac{RT_f^2/E}{T_f - T_0} \right) \right] \\ b_{\text{eff}} &= b_0 \frac{T_0}{T_f} \frac{RT_f^2/E}{T_f - T_0} \end{aligned} \right\} \quad (5)$$

The term T_f was assumed to be the equilibrium flame temperature, which was calculated by the method of reference 6. Low-temperature activation energies of 38 kilocalories per mole for propane (reference 7) and 40 kilocalories per mole for ethylene (reference 8) were used. The transport properties were estimated by extrapolation from tables presented in references 6, 9, and 10 and by the relations:

$$\lambda = (C_p + \frac{5}{4} R) \frac{\eta}{M} \quad (\text{reference 10}) \quad (6)$$

$$\eta = \sum_j \eta_j N_j \quad (7)$$

For combustion products, equation (7) gives values within 1 percent of those obtained by method of reference 11.

$$C_p = \sum_j C_{p_j} N_j \quad (8)$$

$$D = 1.336 \eta / \rho \quad (\text{reference 12}) \quad (9)$$

$$\rho = p / MRT \quad (10)$$

$$Z = \left(\frac{\sigma_1 + \sigma_2}{2} \right)^2 \sqrt{8\pi RT \frac{M_1 + M_2}{M_1 M_2}} \quad (\text{reference 13}) \quad (11)$$

The remaining factor P , used herein as a semiempirical factor, is found by averaging the values calculated by substituting the experimental values of maximum flame velocity for a given fuel and temperature into equation (4). Therefore, the predicted flame velocities discussed hereinafter are actually relative values for particular fuels and temperatures.

The curves for U_{max} against α predicted by this method are compared with the experimental lines in figure 4, and the individual points are given in table III. For propane, the mean deviation of the relative

predictions is approximately 17 percent at 311° K and 12 percent at 422° K; for ethylene, the mean deviation is approximately 4 percent at 311° K and 3 percent at 422° K. The predicted curves show a negative second derivative, $(d^2U_{\max}/d\alpha^2) < 0$, just as the isooctane curves did in reference 1.

Diffusion theory. - The approximate equation given by Tanford and Pease (reference 4), which is based on the diffusion of chain carriers for the oxidation process (active particles) from the flame zone into the unburned gas, may be written

$$U = \sqrt{\frac{L_m Q' n}{Q \theta_m^2} \sum_i \frac{k_i p_i D_m}{B_i}} \quad (12)$$

For these calculations, the active particles are assumed to be hydrogen and oxygen atoms and hydroxyl radicals. The mean combustion-zone temperature is assumed to be $0.7 T_f$ (reference 4); T_f and the active-particle concentrations p_i are calculated by the method of reference 6 on the assumption of adiabatic thermal equilibrium. The diffusion coefficients D_m are calculated by use of the temperature dependence determined from equation (9) and reference values from the Stefan-Maxwell equation (reference 12). The recombination factor B_i is calculated by the method of Tanford (reference 14) for hydrogen atoms and is assumed to be unity for hydroxyl radicals and oxygen atoms. It was assumed that the specific rate constants for all three types of active particles could be represented by a weighted mean specific rate constant k_a . By solving for k_a with the aid of experimental flame velocities in equation (12), it was found that k_a does not exhibit an Arrhenius-type temperature dependence, but is nearly constant. As a result, k_a has been used as an arbitrary constant much as P was in the treatment of the Semenov equation, and the predicted flame velocities are again relative values for a given fuel and temperature.

The flame velocity predictions obtained by this method are presented in figure 5 and table III. The mean deviations for both propane and ethylene are seen to be approximately 5 to 6 percent. All the predicted curves show a negative second derivative.

It is shown in reference 1 that for isooctane-oxygen-nitrogen mixtures no linear correlation between maximum flame velocity and either $(6.5 p_H + p_{OH} + p_O)$ or the square root of this sum held for the entire range of α . Such correlations have been reported when flame velocity was varied by varying initial temperature (reference 2) or equivalence ratio (reference 15). The present data again show no linear correlations that hold for the entire experimental range as maximum flame velocity is varied by varying α ; this is evidenced by the propane data presented in figure 6.

SUMMARY OF RESULTS

A study of the effect of oxygen concentration on the flame velocities of propane and ethylene at two initial temperatures and atmospheric pressure yielded the following results:

1. Maximum flame velocity U_{\max} varied linearly with the mole fraction of oxygen in the primary oxygen-nitrogen mixture α , over the range 0.166 to 0.496 for propane and from 0.166 to 0.349 for ethylene, at both 311° and 422° K, as represented by

$$U_{\max} = A\alpha - B$$

Empirical equations which also include the effect of initial temperature T_0 over the range from 311° to 422° K take the form:

$$U_{\max} = C T_0^m (\alpha - \alpha_x)$$

The least-squares values of the constants A , B , C , m , and α_x for this equation are presented.

2. Subject to the approximations inherent in the theoretical equations and on the assumption that adiabatic thermal and chemical equilibria are attained at the end of the flame zone, it was found that:

a. Relative flame velocities predicted by the Semenov bimolecular equation with activation energies for low-temperature oxidation showed mean deviations from experimental flame velocities of the order of 5 percent for ethylene and 15 percent for propane.

b. With an average specific rate constant, which was assumed to be valid for hydrogen atoms, oxygen atoms, and hydroxyl radicals and to be independent of temperature, the Tanford-Pease equation predicted relative flame velocities with mean deviations from experimental flame velocities of the order of 5 percent for either fuel.

c. Both these equations predicted a decreasing rate of change of maximum flame velocity with increasing oxygen concentrations; this same behavior was previously reported for isooctane.

d. Neither the sum of the effective active particle concentrations nor the square root of this sum was found to be related linearly to maximum flame velocity for the entire range of α investigated.

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TABLE I - EXPERIMENTAL VALUES OF MAXIMUM FLAME VELOCITY OF PROPANE- AND ETHYLENE-OXYGEN-NITROGEN MIXTURES

Fuel	α $\left(\frac{O_2}{O_2+N_2}\right)$	Initial mixture temperature, T_0 ($^{\circ}K$)	Stream-flow Reynolds number	Tube diameter (cm)	Maximum flame velocity (cm/sec)		
					Experimental	Empirical equations (see table II)	
						Least-squares lines $U = A\alpha - B$	$U = CT_0^m(\alpha - \alpha_x)$
Propane	0.166	311	700	1.256	21.6	16.1	19.7
	.210	311	1600	1.256	46.5	42.9	46.0
	.210	311	1600	1.256	43.8	42.9	46.0
	.250	311	1500	.838	63.0	67.3	69.8
	.294	311	1500	.617	89.0	94.1	96.1
	.347	311	1900	.465	120.7	126.3	127.7
	.349	311	1900	.465	126.3	127.6	128.8
	.496	311	1900	.297	222.6	217.1	216.6
	.496	311	1900	.297	217.8	217.1	216.6
	.166	422	650	1.256	37.8	32.7	28.1
	.166	422	650	1.256	36.5	32.7	28.1
	.210	422	900	.838	65.7	68.9	65.5
	.210	422	900	.838	67.9	68.9	65.5
	.250	422	1000	.617	101.3	101.9	99.5
	.294	422	1400	.465	129.5	138.2	136.9
	.347	422	1350	.297	183.4	181.8	182.0
	.349	422	1350	.297	182.8	183.5	183.7
	.496	422	2000	.297	306.3	304.6	308.7
	.496	422	2000	.297	306.6	304.6	308.7
Ethylene	.166	311	1200	1.256	43.2	42.6	46.2
	.166	311	1200	1.256	51.3	42.6	46.2
	.210	311	1400	.617	80.4	81.3	84.6
	.210	311	1400	.617	81.7	81.3	84.6
	.250	311	1900	.465	107.5	116.6	119.5
	.250	311	1900	.465	110.2	116.6	119.5
	.294	311	1500	.297	150.6	155.4	157.9
	.296	311	1500	.297	158.6	157.2	159.6
	.347	311	2000	.297	208.2	202.1	204.1
	.349	311	2000	.297	207.9	203.9	205.8
	.166	422	1000	.838	67.8	69.3	66.3
	.210	422	1200	.617	125.0	121.9	121.3
	.210	422	1200	.617	129.0	121.9	121.3
	.250	422	1200	.297	162.7	169.7	171.3
	.250	422	1200	.297	161.8	169.7	171.3
	.294	422	1500	.297	221.2	222.3	226.3
	.296	422	1500	.297	232.0	224.7	228.8
	.347	422	2200	.297	282.5	285.7	292.5
	.349	422	2200	.297	291.5	288.1	295.0

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TABLE II - EMPIRICAL EQUATIONS AND CONSTANTS FOR ISOCTANE-,
PROPANE-, AND ETHYLENE-OXYGEN-NITROGEN MIXTURES

Fuel	Equation (2): $U_{max} = A\alpha - B$ Equation (3): $U_{max} = CT_0^m (\alpha - \alpha_x)$						
	Constants						
	A at 311° K	B at 311° K	A at 422° K	B at 422° K	C	m	α_x
Isooctane ^a	409	51.1	601	69.3	0.133	1.40	0.120
Propane	609	85.0	824	104.1	.766	1.16	.133
Ethylene	882	103.8	1196	129.3	.998	1.18	.113

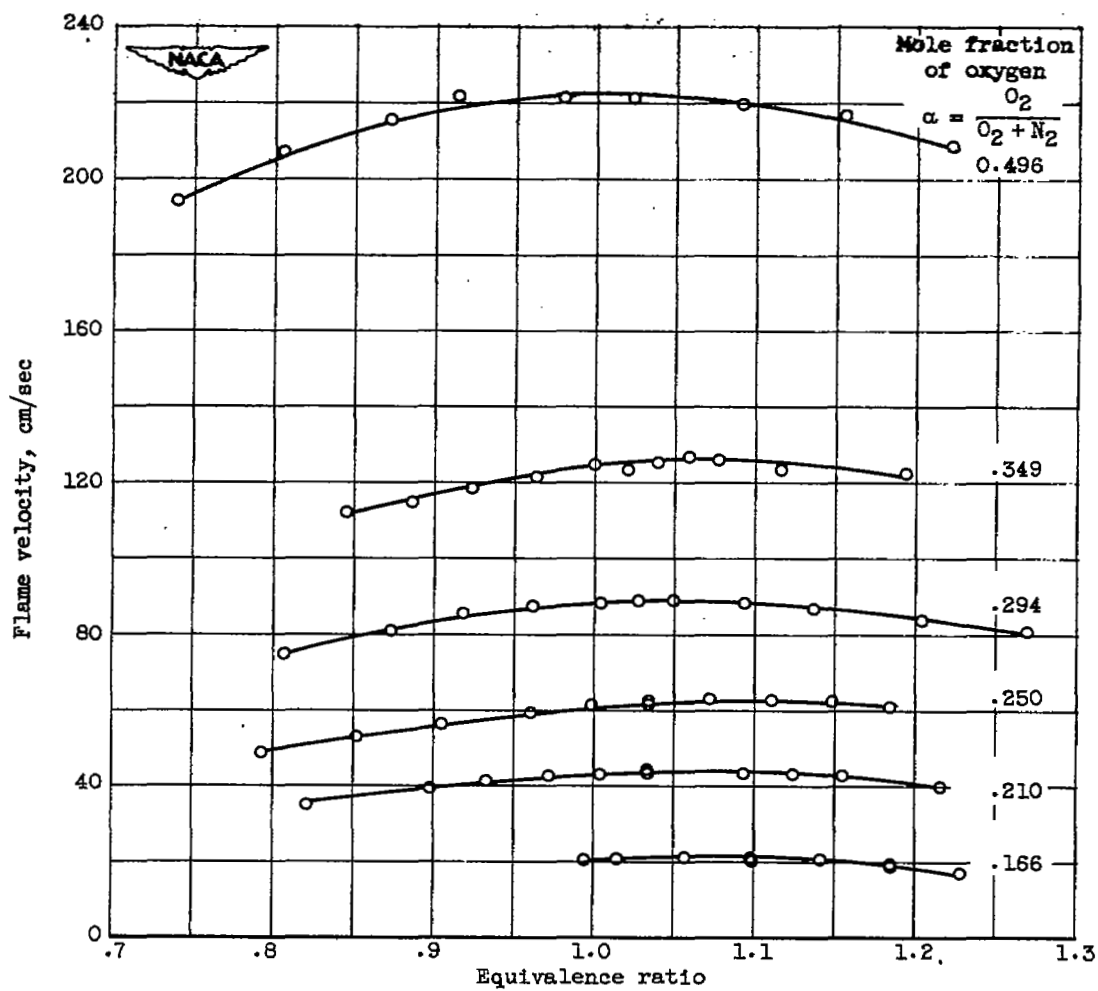
^aReference 1.



TABLE III - THEORETICALLY PREDICTED VALUES OF RELATIVE MAXIMUM FLAME VELOCITY OF
PROPANE- AND ETHYLENE-OXYGEN-NITROGEN MIXTURES

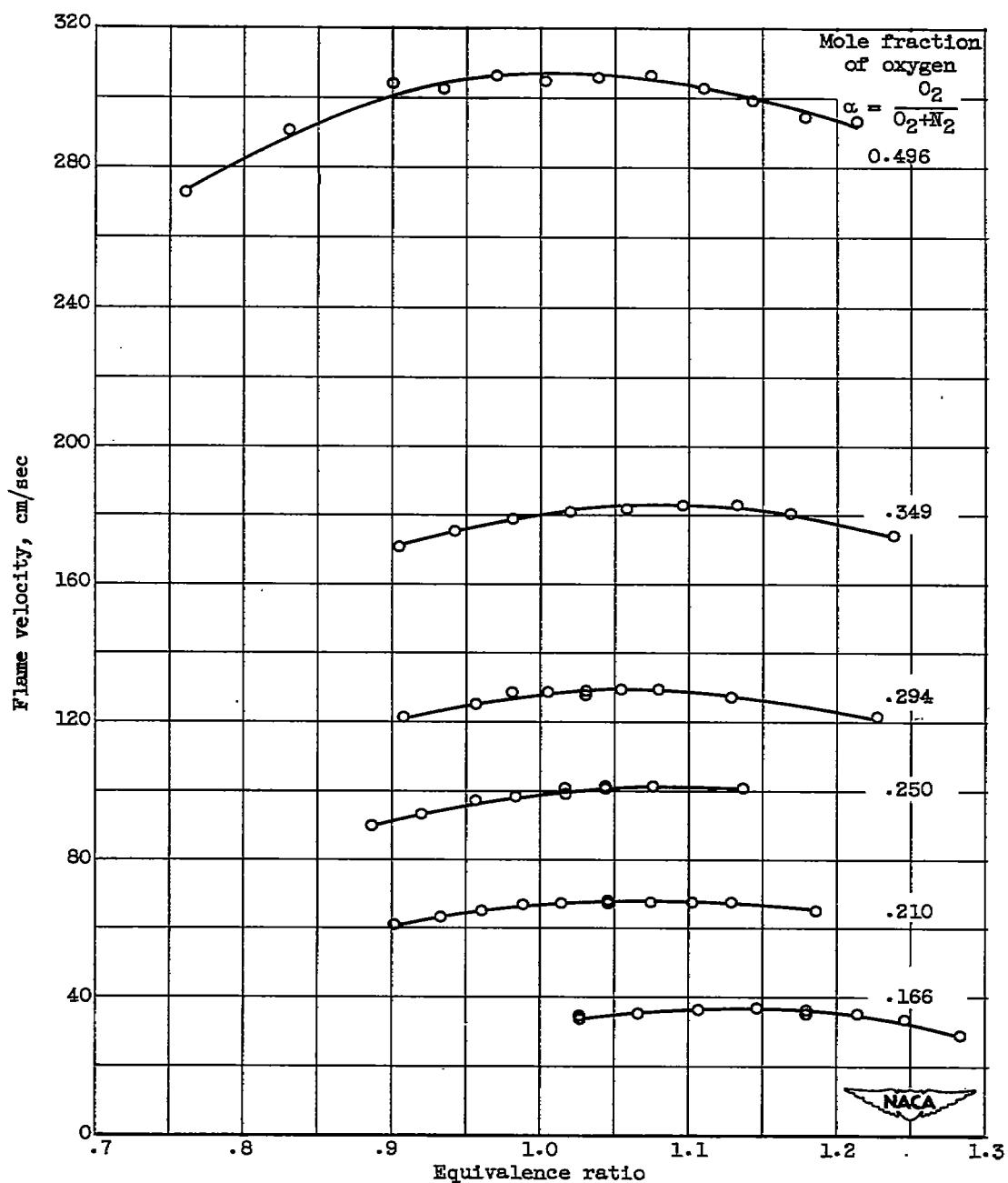


Fuel	α $\left(\frac{O_2}{O_2+N_2}\right)$	Initial mixture tempera- ture, T_0 (°K)	Flame tempera- ture, T_f (°K)	EQUIVA- LENCE RATIO ϕ	Equilibrium radical concentrations (mole fraction $\times 10^3$)			Maximum flame velocity (cm/sec)		
					P_H	P_{OH}	P_O	Semenov equation	Tanford- Pease equation	Experi- mental least squares
Propane	0.166	311	2009	1.06	0.117	0.270	0.006	23.7	17.5	16.1
	.210	311	2293	1.06	.755	2.50	.247	50.9	46.0	42.9
	.250	311	2441	1.04	1.78	6.10	1.04	70.5	70.0	67.3
	.294	311	2561	1.03	3.61	11.03	2.59	91.7	95.6	94.1
	.348	311	2670	1.03	6.75	17.60	5.18	118.1	125.1	127.0
	.496	311	2846	1.00	16.45	36.23	15.11	168.1	184.9	217.1
	.166	422	2092	1.06	.198	.558	.023	41.0	32.8	32.7
	.210	422	2346	1.06	1.03	3.33	.403	79.5	73.7	68.9
	.250	422	2483	1.05	2.33	7.10	1.34	108.8	107.9	101.9
	.294	422	2595	1.04	4.44	12.24	3.08	138.5	143.4	138.2
	.348	422	2698	1.04	7.89	18.94	5.86	175.1	184.0	182.7
	.496	422	2864	1.00	17.84	37.74	16.32	236.8	265.0	304.6
Ethylene	.166	311	2100	1.16	.315	.336	.009	41.5	38.0	42.6
	.210	311	2401	1.14	1.68	3.24	.407	88.7	85.8	81.3
	.250	311	2550	1.10	3.42	7.92	1.79	118.5	122.6	116.6
	.295	311	2665	1.08	6.20	13.78	4.25	152.8	160.9	156.3
	.348	311	2766	1.08	10.54	20.79	7.87	194.2	203.2	203.0
	.166	422	2183	1.15	.492	.668	.031	67.7	62.2	69.3
	.210	422	2452	1.13	2.12	4.36	.690	128.9	127.4	121.9
	.250	422	2587	1.11	4.23	9.04	2.23	173.2	178.0	169.7
	.295	422	2697	1.10	7.50	14.92	4.84	220.9	230.8	223.5
	.348	422	2794	1.10	12.28	21.99	8.63	277.1	287.8	286.9



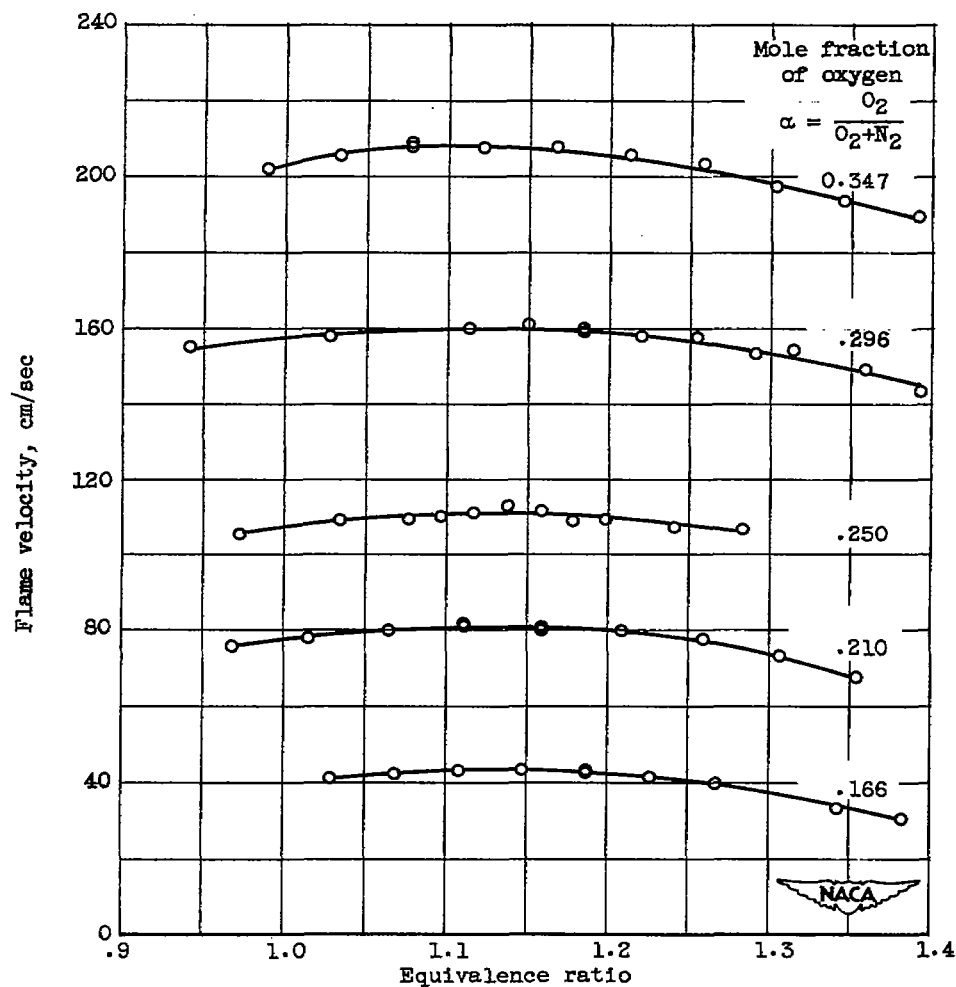
(a) Propane-oxygen-nitrogen mixtures at 311° K.

Figure 2. - Flame velocity as function of equivalence ratio over range of oxygen concentrations.



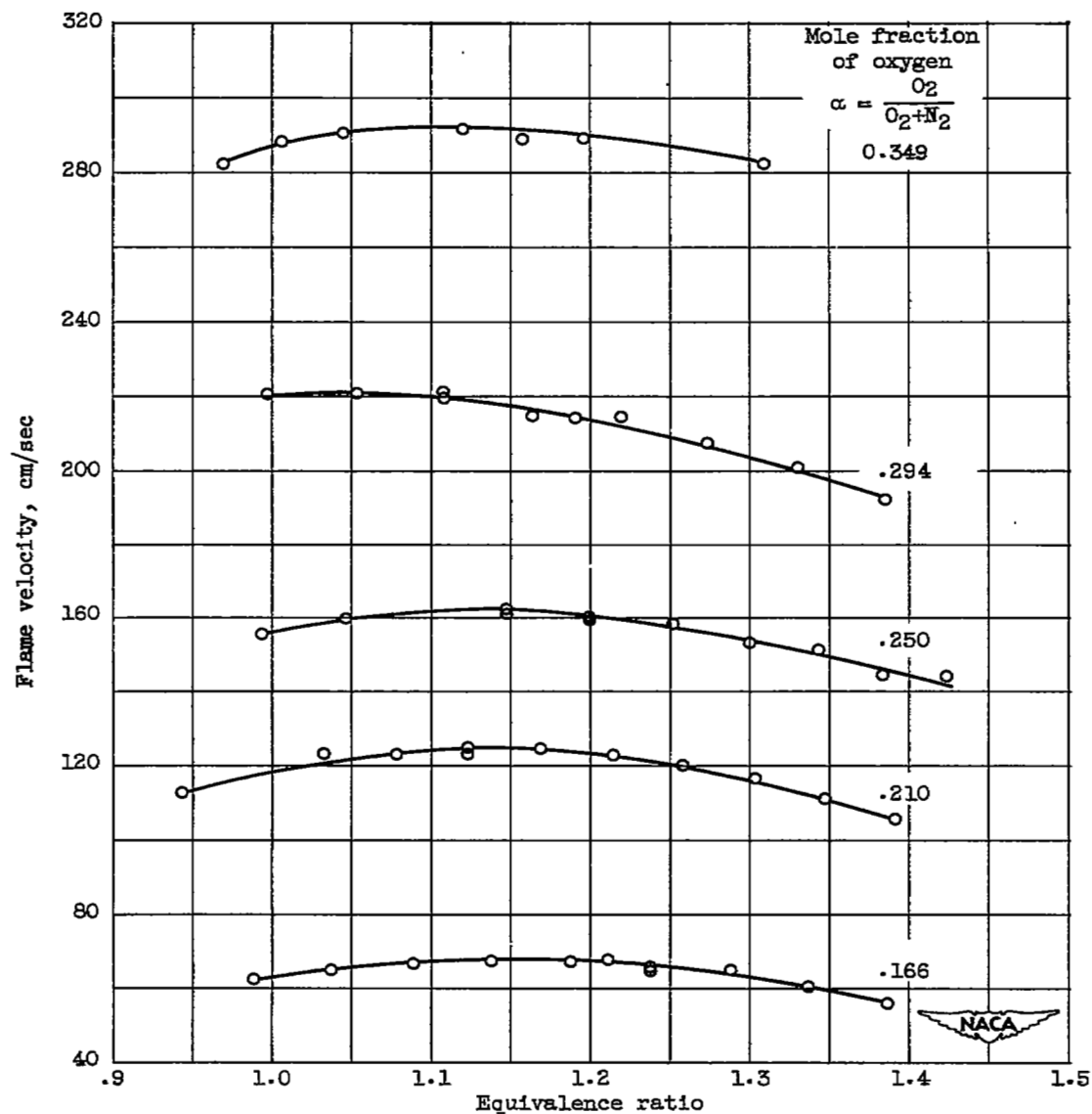
(b) Propane-oxygen-nitrogen mixtures at 422° K.

Figure 2. - Continued. Flame velocity as function of equivalence ratio over range of oxygen concentrations.



(c) Ethylene-oxygen-nitrogen mixtures at 311° K.

Figure 2. - Continued. Flame velocity as function of equivalence ratio over range of oxygen concentrations.



(d) Ethylene-oxygen-nitrogen mixtures at 422° K.

Figure 2. - Concluded. Flame velocity as function of equivalence ratio over range of oxygen concentrations.

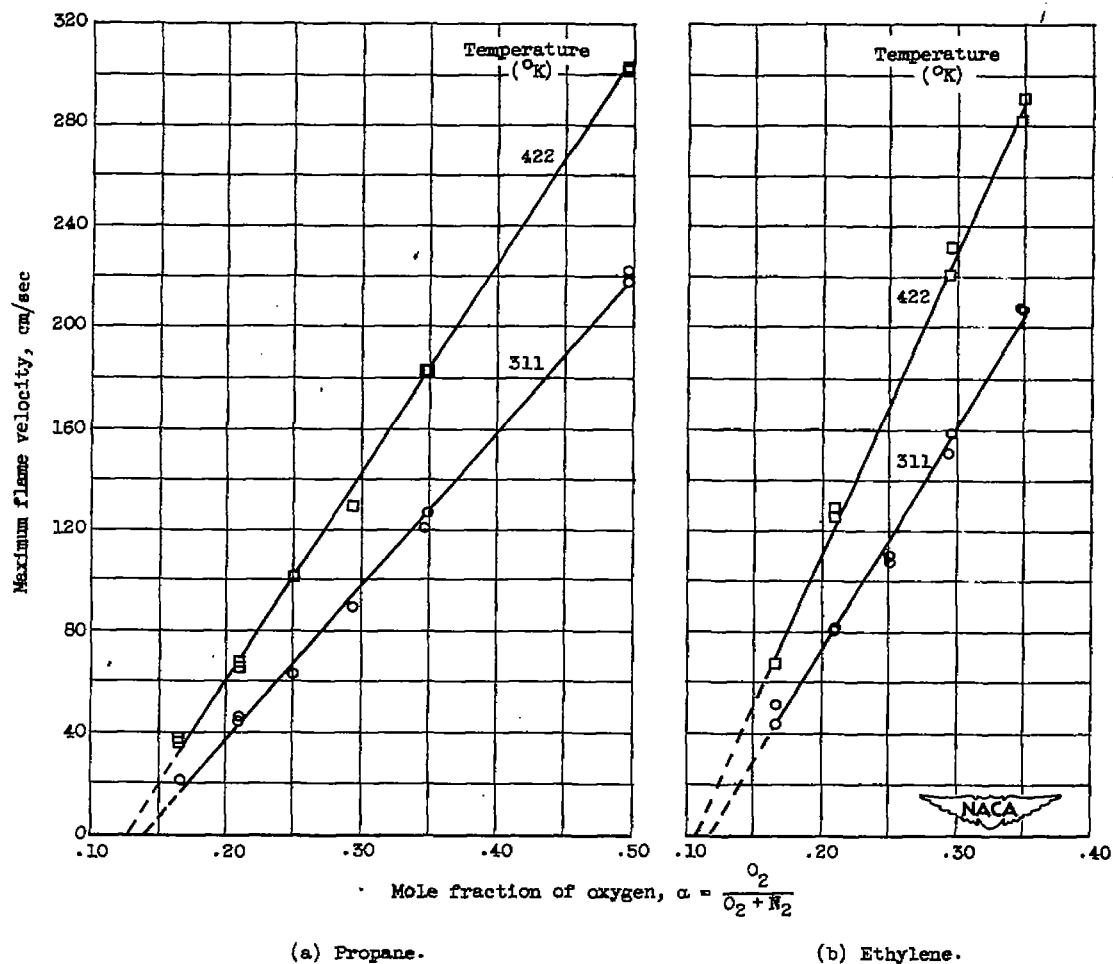


Figure 3. - Effect of oxygen concentration on maximum flame velocities of propane- and ethylene-oxygen-nitrogen mixtures at 311° and 422° K.

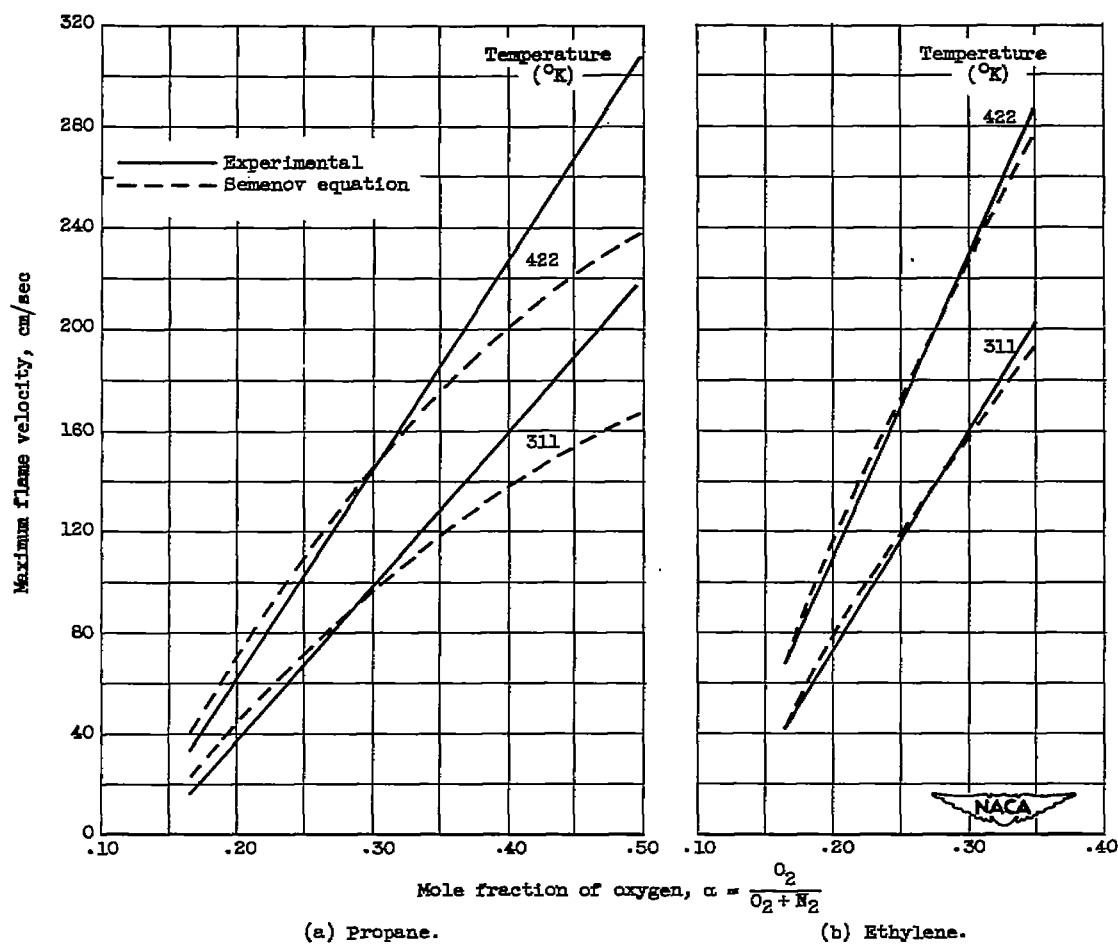


Figure 4. - Comparison of curves predicted by Semenov bimolecular equation with experimental results.

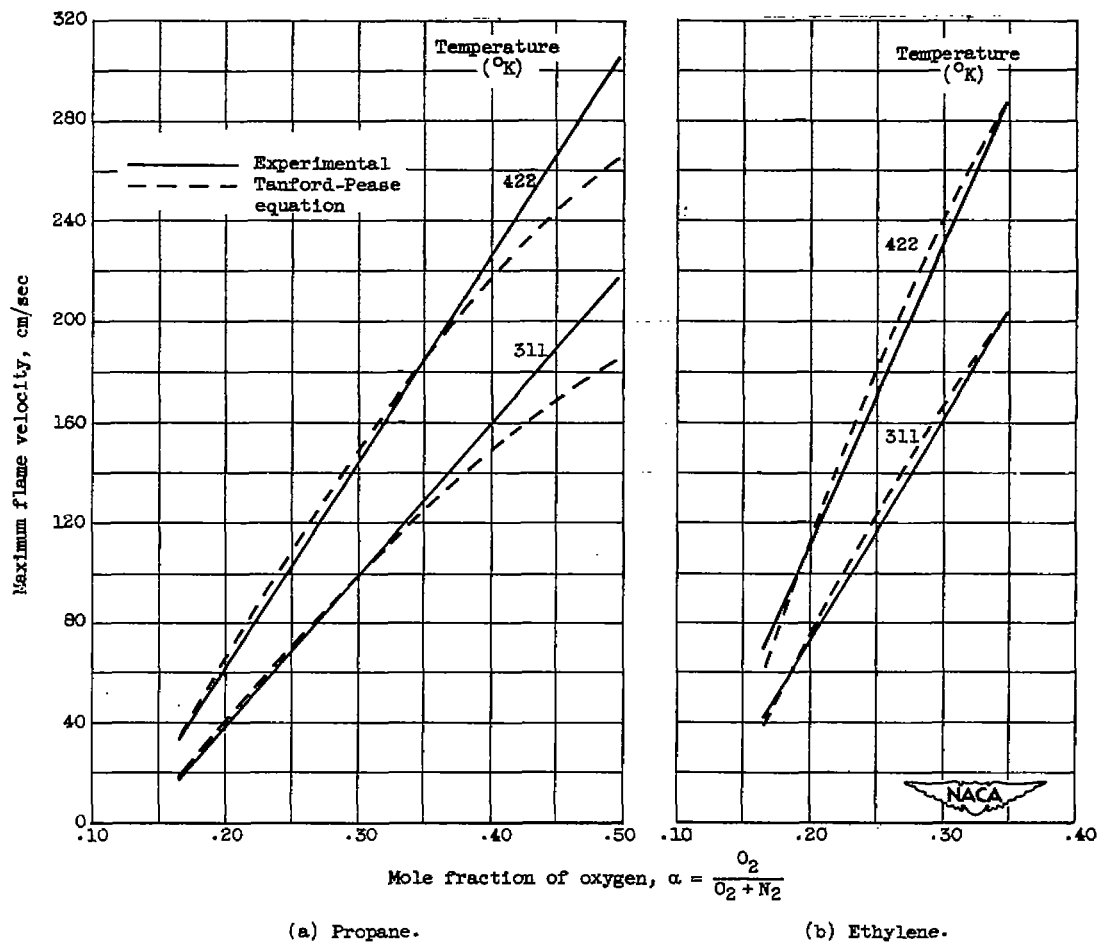


Figure 5. - Comparison of curves predicted by Tanford-Pease theory with experimental results.

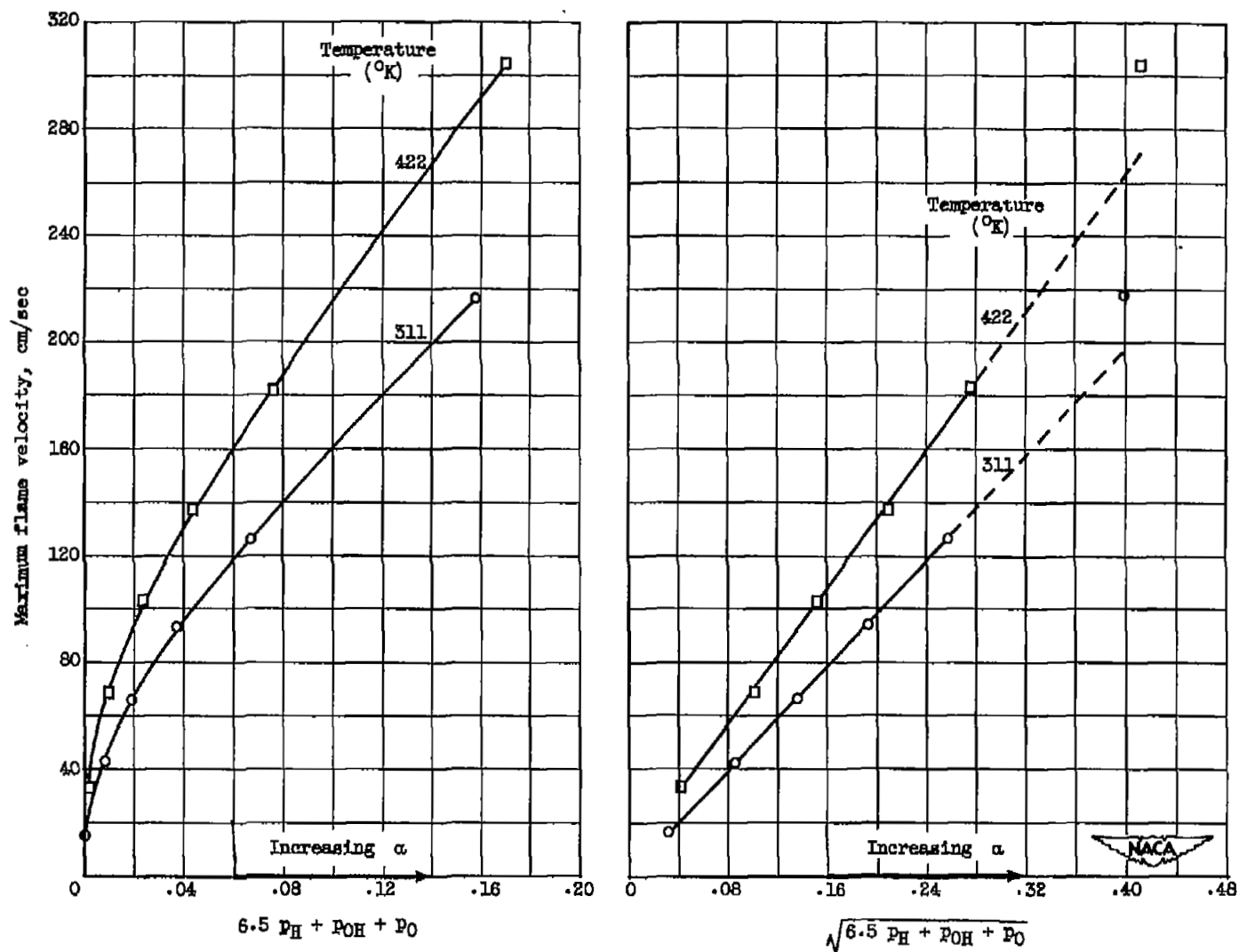


Figure 8. - Relation between maximum flame velocity of propane-oxygen-nitrogen mixtures and effective active-particle concentrations.

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